Chapter IV:
Radioactive decay

Summary

1. Law of radioactive decay
2. Decay chain/Radioactive filiation
3. Quantum description
4. Types of radioactive decay

History

- Radioactivity was discover in 1896 by Henri Becquerel while working on uranium salts
- On Earth some elements show « natural » radioactivity due to their lifetime comparable to the age of the Earth (principally uranium and thorium)
- Artificial radioactivity (artificial production radioactive elements) is possible with use of particles accelerators or nuclear reactors → first time (Irène and Frédéric Jolliot-Curie):

$$
\begin{aligned}
{}^{27}_{13}\mathrm{Al} + {}^{4}_{2}\alpha &\rightarrow + {}^{30}_{15}\mathrm{P} + n \\
{}^{30}_{15}\mathrm{P} &\rightarrow + {}^{30}_{14}\mathrm{Si} + \beta^{+} + \nu_e
\end{aligned}
$$

Law of radioactive decay (1)

- Radioactive decay has a statistical character → impossible to precisely predict when a disintegration will happen → only probabilities may be given
- If 4 conditions are fulfilled:
  1. atoms are identical
  2. they are independent
  3. their mean life is long
  4. their number is large

  → the Poisson statistic can be applied:

$$P_x(T) = \frac{(\lambda T)^x}{x!} e^{-\lambda T}$$

- P_x is the probability for x disintegrations in a time interval T and λ is the probability of disintegration per unit time = decay (desintegration) constant (independent of the nucleus age)

Law of radioactive decay (2)

- λdt is the disintegration probability of a nucleus in the time interval *dt*
- The survival probability of a nucleus at time t (if existing in $t = 0$) →

$$P_0(t) = e^{-\lambda t}$$

- If N_0 is the initial number (at $t = 0$) of nuclei → the number of survival nuclei *N(t)* at time *t* is:

$$N(t) = N_0 e^{-\lambda t}$$

Half-life and activity (1)

- The half-life $T_{½}$ is time is time taken for half the radionuclide's atoms to decay →

$$\begin{aligned} N(T_{1/2}) &= \frac{N_0}{2} = N_0 e^{-T_{1/2}\lambda} \\ &\Rightarrow \\ T_{1/2} &= \frac{\ln 2}{\lambda} \end{aligned}$$

- The mean lifetime for decaying atoms τ is defined as the arithmetic mean of all the atoms' lifetimes →

$$\tau = \frac{\int_0^\infty t \left|\frac{dN}{dt}\right| dt}{\int_0^\infty \left|\frac{dN}{dt}\right| dt} = \frac{\int_0^\infty \lambda t e^{-\lambda t}}{\int_0^\infty \lambda e^{-\lambda t}} = \frac{1}{\lambda}$$

Half-life and activity (2)

- Activity *A(t)* at time *t* is defined as the mean number of disintegrations per time unit →

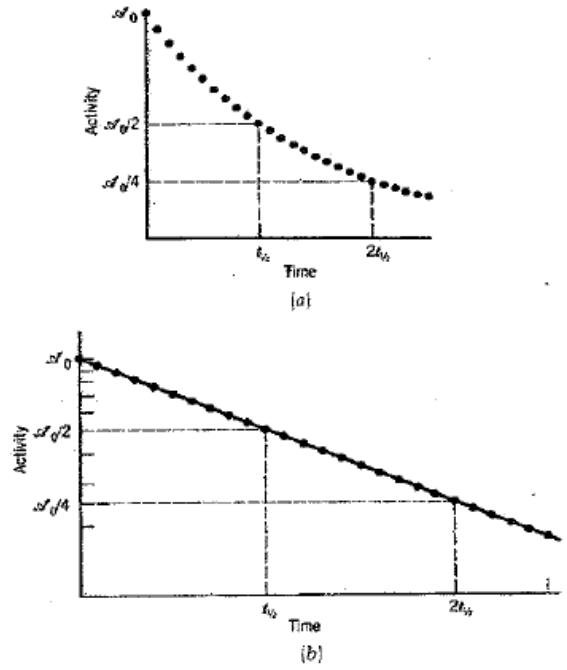
$$A(t) \equiv \lambda N(t) = \left| \frac{dN}{dt} \right|$$

- The activity unit is Becquerel (Bq) → 1 Bq = 1 disintegration per second (old unit → Curie (Ci) corresponding to the activity of 1 g of ^{226}Ra → 1 Ci = 3.7×10^{10} Bq)

Radioactive filiation (1)

- Simple case: Radioactive nucleus 1 (N_0 at time t = 0) decays with decay constant λ_1 to stable nucleus 2 →

$$N_1(t) = N_0 e^{-\lambda_1 t}$$
$$N_2(t) = N_0 \left(1 - e^{-\lambda_1 t}\right)$$



Radioactive filiation (2)

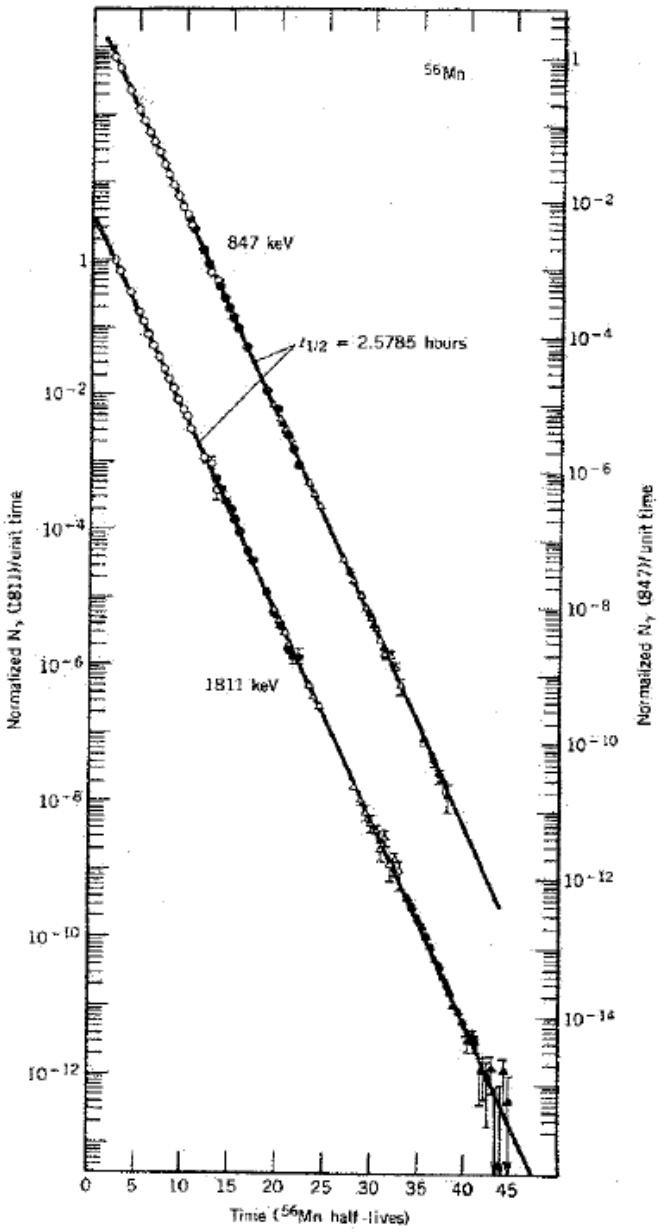
- Two decay modes are sometimes possible → λ_a and λ_b
- Total decay rate:

$$-\left(\frac{dN}{dt}\right) = -\left(\frac{dN}{dt}\right)_a - \left(\frac{dN}{dt}\right)_b = (\lambda_a + \lambda_b)N = \lambda_t N$$

- The total decay constant is $\lambda_a + \lambda_b = \lambda_t$
- Practically λ_t is observed while λ_a and λ_b are determined by the final number of isotopes of each mode:

$$\begin{aligned} N_1(t) &= N_0 e^{-\lambda_t t} \\ N_{2,a}(t) &= \frac{\lambda_a}{\lambda_t} N_0 \left(1 - e^{-\lambda_t t}\right) \\ N_{2,b}(t) &= \frac{\lambda_b}{\lambda_t} N_0 \left(1 - e^{-\lambda_t t}\right) \end{aligned}$$

Radioactive filiation (3)



Measurement of the
2 γ rays of ^{56}Mn

Radioactive filiation (4)

- We suppose now $X_1 \overset{\lambda_1}{\rightarrow} X_2 \overset{\lambda_2}{\rightarrow} X_3$
- The number of X_1 (« parent ») decreases following an exponential equation →

$$\frac{dN_1}{dt} = -\lambda_1 N_1 \rightarrow N_1(t) = N_1(0)e^{-\lambda_1 t}$$

- The number of X_2 (« daughter ») increases due to disintegration of X_1 and disintegrates with the disintegration constant λ_2 →

$$\frac{dN_2}{dt} = -\lambda_2 N_2 + \lambda_1 N_1 = -\lambda_2 N_2 + \lambda_1 N_1(0)e^{-\lambda_1 t}$$

- The solution is→

$$N_2(t) = N_2(0)e^{-\lambda_2 t} + \frac{\lambda_1}{\lambda_2 - \lambda_1} N_1(0)\left(e^{-\lambda_1 t} - e^{-\lambda_2 t}\right)$$

Radioactive filiation (5)

- The number of X_3 changes as

$$\frac{dN_3}{dt} = \lambda_2 N_2$$

$$N_3(t) = N_3(0) + N_2(0)\left(1 - e^{-\lambda_2 t}\right) + N_1(0)\left(1 - \frac{\lambda_2 e^{-\lambda_1 t} - \lambda_1 e^{-\lambda_2 t}}{\lambda_1 - \lambda_2}\right)$$

- Practically → measures of activities $A_1 = \lambda_1 N_1$ and $A_2 = \lambda_2 N_2$ → assuming $N_2(0) = N_3(0) = 0$ →

$$A_1(t) = A_1(0)e^{-\lambda_1 t} \qquad \text{and} \qquad A_2(t) = \frac{\lambda_2}{\lambda_2 - \lambda_1} A_1(0)\left(e^{-\lambda_1 t} - e^{-\lambda_2 t}\right)$$

Equilibria (1)

- We note that $A_1(t)$ is maximum at $t = 0$ and zero at $t = \infty$ and that $A_2(t)$ is zero at $t = 0$ and $t = \infty \rightarrow A_2(t)$ has a maximum for $dA_2(t)/dt = 0 \rightarrow$

$$\frac{d(A_2)}{dt} = 0 = -\lambda_1 e^{-\lambda_1 t_m} + \lambda_2 e^{-\lambda_2 t_m}$$

$$\Rightarrow \quad t_m = \frac{\ln \lambda_2/\lambda_1}{\lambda_2 - \lambda_1}$$

- This maximum happens when the activities of parent and daughter are equal $\rightarrow A_1(t_m) = A_2(t_m)$

$$e^{-\lambda_1 t_m} = \frac{\lambda_2}{\lambda_2 - \lambda_1}\left(e^{-\lambda_1 t_m} - e^{-\lambda_2 t_m}\right)$$

$$\Rightarrow \quad t_m = \frac{\ln \lambda_2/\lambda_1}{\lambda_2 - \lambda_1}$$

Equilibria (2)

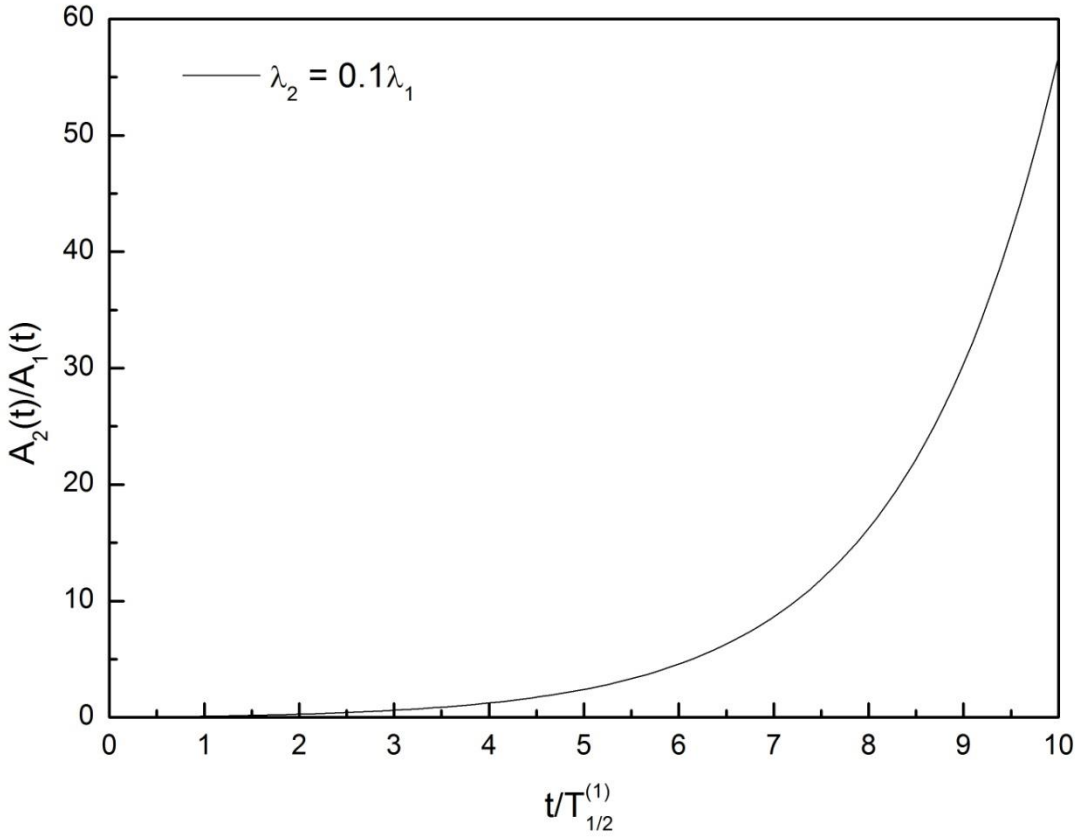
- At t_m → we have « *ideal equilibrium*»
- The ratio of activities of X_2 and X_1 is →

$$\frac{A_2(t)}{A_1(t)} = \frac{\lambda_2}{\lambda_2 - \lambda_1}\left(1 - e^{-(\lambda_2 - \lambda_1)t}\right)$$

- For $t < t_m$ → always $A_1 > A_2$
- For $t > t_m$ → always $A_1 < A_2$
- The specific relation between parent and daughter depends on the relative values of their disintegration constants→ 3 cases →
  1. $\lambda_2 < \lambda_1$
  2. $\lambda_2 > \lambda_1$
  3. $\lambda_2 \gg \lambda_1$

Non-equilibrium: $\lambda_2 < \lambda_1$

- X_1 isotopes disintegrate faster than filiation products X_2 → the ratio of activities increases without limit

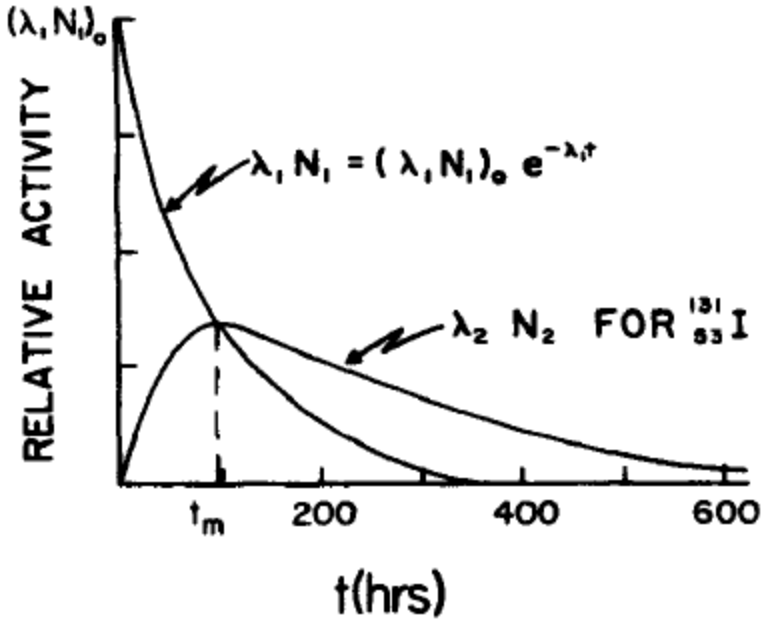


Example with $\lambda_2 < \lambda_1$

- Disintegration of metastable tellurium →

$$^{131m}_{52}\mathrm{Te} \xrightarrow{T_{1/2} = 30h} {}^{131}_{53}\mathrm{I} \xrightarrow{T_{1/2} = 193h} {}^{131}_{54}\mathrm{Xe}$$

- We have thus → λ_1 = 2.31 10^{-2} h^{-1} and λ_2 = 3.59 10^{-3} h^{-1}

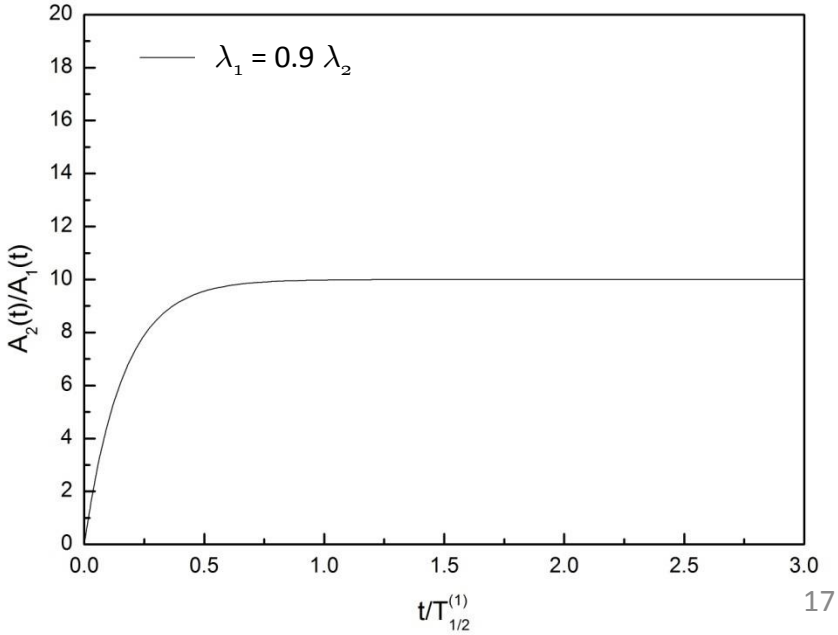


Transient equilibrium: $\lambda_2 > \lambda_1$

- The activities ratio increases as a function of time and reaches a constant value → for $t \to \infty$:

$$\frac{A_2(t)}{A_1(t)} \simeq \frac{\lambda_2}{\lambda_2 - \lambda_1}$$

- The daughter activity decreases at the same rate as that of the parent → this equilibrium is called transient equilibrium



Secular equilibrium: $\lambda_2 \gg \lambda_1$

- The activities ratio increases as a function of the time and reaches 1 pour $t \rightarrow \infty$:

$$\frac{A_2(t)}{A_1(t)} \simeq 1$$

- The parent and daughter activities become equal → secular equilibrium
- Example → disintegration of radium →

$$^{226}_{88}\mathrm{Ra} \xrightarrow{T_{1/2} = 1602 \text{ an}} {}^{222}_{86}\mathrm{Rn} \xrightarrow{T_{1/2} = 3.8 \text{ j}} {}^{218}_{84}\mathrm{Po}$$

- We have → λ_1 = 1.18 10^{-6} j^{-1} and λ_2 = 1.81 10^{-1} j^{-1}

Bateman equations

- We consider $X_1 \xrightarrow{\lambda_1} X_2 \xrightarrow{\lambda_2} X_3 \rightarrow \ldots$
- Generalization of previous equations →

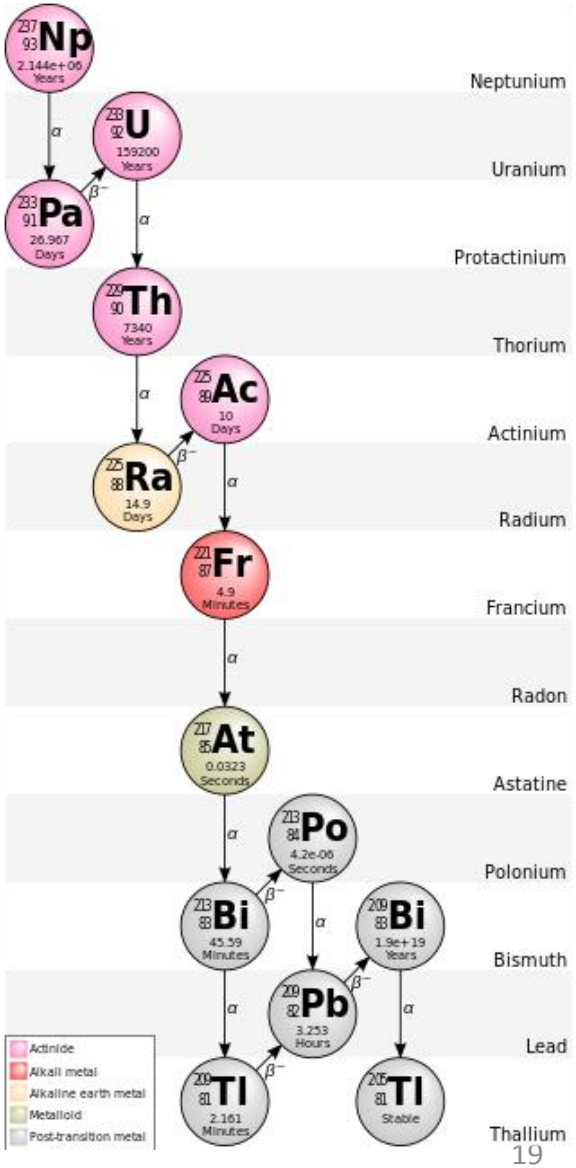
$$dN_i = \lambda_{i-1} N_{i-1} dt - \lambda_i N_i dt$$

- General solution for N_0 nuclei of type 1 and none for other types is given by the Bateman equations →

$$A_n = N_0 \sum_{i=1}^{n} c_i e^{-\lambda_i t}$$

$$\text{where } c_m = \frac{\prod_{i=1}^{n} \lambda_i}{\prod{}'_{i=1}^{n} (\lambda_i - \lambda_m)}$$

- ' means that term with $i = m$ is omitted
- Secular equilibrium is possible ($A_1 = A_2 = \ldots$)



ORIGEN

- Bateman formula can be implemented easily in computer code → but if $\lambda_p \approx \lambda_j$ for some isotope pair → cancellation can lead to computational errors → other methods such as numerical integration or the matrix exponential method are in use
- ORIGEN (Oak Ridge Isotope GENeration in SCALE) code calculates the decay chains by the matrix exponential method
- ORIGEN was developed for the Nuclear Regulatory Commission and the Department of Energy (USA) → easy-to-use standardized method of isotope depletion/decay analysis for spent fuel, fissile material and radioactive material
- It can be used to solve for spent fuel characterization, isotopic inventory, radiation source terms and decay heat

Application: Production of radioelements (1)

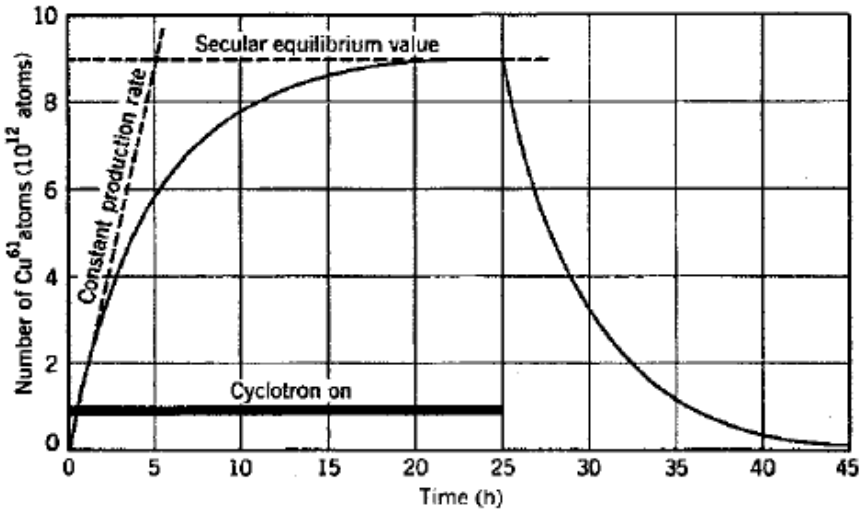
- Stable element placed into a reactor or an accelerator (such a cyclotron) → nucleus captures a neutron or a charged particle → possible production of a radioelement
- The production rate R (unit: $m^{-3}s^{-1}$) depends on the target atom density N_0 (unit: m^{-3}), on the density of current J of the beam (unit: $m^{-2}s^{-1}$) and on the reaction cross section σ unit: m^2) → $R = N_0 \sigma J$
- As $\sigma \simeq 10^{-24}$ cm^2 and $J \simeq 10^{14}$ $cm^{-2}s^{-1}$ → the probability to convert a stable particle is $\simeq 10^{-10}$ s^{-1} → the number of converted particle is small → the number of target nuclei is constant → R is constant

Application: Production of radioelements (2)

- We consider: $N_0 \xrightarrow{R} N_1 \xrightarrow{\lambda_1} N_2$
- We have thus:

$$dN_1 = Rdt - \lambda_1 N_1 dt \rightarrow N_1(t) = \frac{R}{\lambda_1}(1 - e^{-\lambda_1 t}) \rightarrow A_1 = R(1 - e^{-\lambda_1 t})$$

- If the irradiation time T $\ll T_{½} \rightarrow A_1 = R\lambda_1 T$
- If the irradiation time T $\gg T_{½} \rightarrow A_1 = R$ (secular equilibrium)



Production ^{61}Cu ($T_{½}$ = 3.4 h) due to bombardment of ^{61}Ni by deuteron → use for positron emission tomography (PET)

Application: Carbon-14 dating

- Radiocarbon (14-C) is constantly being created in the atmosphere by the interaction of cosmic rays with atmosphere:

$$^{14}_{7}\mathrm{N} + ^{1}_{0}n \rightarrow ^{14}_{6}\mathrm{C} + ^{1}_{1}\mathrm{H}$$

- The resulting radiocarbon is incorporated into plants by photosynthesis → into animals by eating the plants
- During its life → a plant or animal is exchanging carbon with its surroundings → same proportion of 14-C as the biosphere
- When dying → no more 14-C acquiring and decay of 14-C in the organic sample
- The measurement of the ratio $^{14}C/C_{total}$ gives the sample age
- As $T_{½}$ = 5730 ± 40 years → dating is possible for age between a few hundred years and about 50000 years

Quantum description of radioactive decays (1)

- Solving the Schrödinger equation for various time-independent potentials → energy levels are *stationary* states
- A system in a particular stationary state will remain in that state for all times → no transition → no decay
- If we assume one state being the mixture of two (or more) states → $\psi = c_1\psi_1 + c_2\psi_2$ → probability $|c_1|^2$ to be found in 1 and $|c_2|^2$ to be found in 2 → for time-independent potential → c_1 and c_2 are independent on time → ≠ with observation
- We are forced to abandon the notion of pure states with well-defined wave-functions → difficult interpretation of nuclear structure

Quantum description of radioactive decays (2)

- We assume a potential of the form *V + V'* where *V* is the nuclear potential that gives stationary states and *V'* is a very weak additional potential that causes the transition between the states
- Neglecting *V'* → we obtain the static nuclear wave functions
- These wave functions are used to calculate the transition probability between the « stationary states » under the influence of *V'* → this transition probability is λ
- Fermi Golden Rule →

$$\lambda = \frac{2\pi}{\hbar}|V'_{fi}|^2\rho(E_f) \text{ where } V'_{fi} = \int \psi_f^* V' \psi_i d\boldsymbol{r}$$

Quantum description of radioactive decays (3)

- The potential *V'* depends on the particular type of transition which is considered

- The transition probability is thus influenced by the density of final states $\rho(E_f)$ → within an energy interval dE_f the number of states accessible to the system is $dn_f = \rho(E_f)dE_f$ → the transition probability is larger if the number of final states accessible for the decay is large

Width of the states (1)

- Solving the Schrödinger equation for time-independent potential V → stationary states of the nucleus $\psi_i(\mathbf{r})$ → the time-dependent wave function $\Psi_i(\mathbf{r},t)$ is

$$\Psi_i(\boldsymbol{r},t) = \psi_i(\boldsymbol{r})e^{-iE_it/\hbar}$$

  where E_i is the energy of the state
- The probability of finding the system in the state is $|\Psi_i(\mathbf{r},t)|^2$ → independent on time for stationary state
- To be consistent with the radioactive decay law we have to introduce the decrease with time exp(-t/τ_i) with $\tau_i = 1/\lambda_i$ →

$$|\Psi_i(t)|^2 = |\Psi_i(t=0)|^2 e^{-t/\tau_i}$$

- The expression of $\Psi_i(\mathbf{r},t)$ becomes →

$$\Psi_i(\boldsymbol{r},t) = \psi_i(\boldsymbol{r})e^{-iE_it/\hbar}e^{-t/2\tau_i}$$

Width of the states (2)

- The resonant state (non-stationary state) can be written →

$$\Psi_i(\boldsymbol{r},t) = \psi_i(\boldsymbol{r}) \exp\left(\frac{-it}{\hbar}(E_i - \frac{i\lambda\hbar}{2})\right)$$

- Complex energy: $E_i - i\lambda\hbar/2$
- Alternatively the state has no a definite energy → the wave function is a superposition of components having different energies (with *A(E)* the probability amplitude to find the state at energy *E*) →

$$\exp\left(\frac{-it}{\hbar}(E_i - \frac{i\lambda\hbar}{2})\right) = \int A(E) \exp\left(\frac{itE}{\hbar}\right)$$

Width of the states (3)

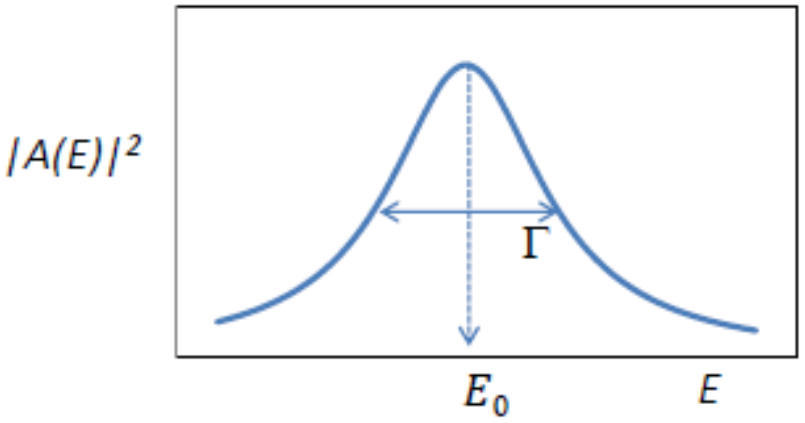
- The probability for finding the state at energy E_i is given by the absolute square of the amplitude →

$$|A(E)|^2 = \frac{1}{4\pi^2} \frac{1}{(E - E_i)^2 + \Gamma_i^2/4}$$

- The shape of such a distribution is *Lorentzian* and $\Gamma_i = \hbar/\tau_i$ is the width of the state *i* → full width at half maximum (FWHM) of such a distribution
- The width is the measure of our inability to determine precisely the energy of the state → it is not a question of instrumental uncertainty

Width of the states (4)

- Another way to understand it → $\Delta E \Delta t \geq \hbar/2$ → if $\Delta t \to \infty$ we can precisely determine the energy of the state because $\Delta E = 0$
- If the state lives on a average for a time τ → we cannot determine its energy except to within an uncertainty of $\Delta E \sim \hbar/\tau$



- It is always possible to speak of transitions between distinct levels because the widths Γ of nuclear levels (typically $\Gamma < 10^{-10}$ MeV) is small compared with their energy spacing ($\sim 10^{-3}$ MeV)

Types of radioactive decay

- There are 3 principal types of decay: α-, β- and γ-decay processes
- In α and β processes → an unstable nucleus emits an α or a β particle as it tries to become a more stable nucleus
- In γ-decay process → an excited state decays toward the ground state without changing the nuclear species

α-decay

- The nucleus emit an α particle i.e. a nucleus of helium: ${}^{4}_{2}\mathrm{He}_{2}$
- The ^{4}He nucleus is a tightly bound system → the kinetic energy released is maximized
- The decay process is

$$ {}^{A}_{Z}X_{N} \rightarrow {}^{A-4}_{Z-2}X'_{N-2} + {}^{4}_{2}\mathrm{He}_{2} $$

- The number of protons and neutrons are separately conserved
- Example (with $T_{½}$ = 1600 years and $E_{kin}(\alpha)$ = 4.8 MeV):

$$ {}^{226}_{88}\mathrm{Ra}_{138} \rightarrow {}^{222}_{86}\mathrm{Rn}_{136} + {}^{4}_{2}\mathrm{He}_{2} $$

β-decay (1)

- The nucleus can correct a proton or a neutron excess by directly converting a proton into a neutron or a neutron into a proton
- Three possible ways for this process → each of them involves another charged particle to conserve electric charge and a (anti-)neutrino to conserve the electronic lepton number
  - β^- decay: $n \rightarrow p + e^- + \overline{\nu}$
  - β^+ decay: $p \rightarrow n + e^+ + \nu$
  - electron capture (ϵ): $p + e^- \rightarrow n + \nu$
- For the electron capture → an atomic electron too close to the nucleus is swallowed
- In all cases where β^+-decay is allowed energetically → electron capture is allowed (competing process) but not the opposite

β-decay (2)

- In β^-- and β^+-decays → a particle is created (electron - « negatron » - and positron, respectively) → they did not exist inside the nucleus before the decay → in contrast with α-decay in which the emitted nucleon were inside the nucleus before the decay
- We also note that the emitted β^- and β^+ show an energy spectrum → the total energy is shared between the 3 bodies
- In electron capture → the neutrino energy is fixed
- Examples:

$$
\begin{aligned}
{}^{131}_{53}\mathrm{I}_{78} &\rightarrow {}^{131}_{54}\mathrm{Xe}_{77} + \beta^- + \bar{\nu} \\
{}^{25}_{13}\mathrm{Al}_{12} &\rightarrow {}^{25}_{12}\mathrm{Mg}_{13} + \beta^+ + \nu \\
{}^{54}_{25}\mathrm{Mn}_{29} + e^- &\rightarrow {}^{54}_{24}\mathrm{Cr}_{30} + \nu
\end{aligned}
$$

- In these processes → *Z* and *N* each change by one unit but *Z + N* = constant

γ-decay

- An excited state decays to a lower excited state or possibly the ground state by emission of a photon of γ radiation with energy equal to the difference between the nuclear states (less a usually negligible correction due to the recoil)
- γ-decay is observed in all nuclei that have excited bound states ($A > 5$) and generally follows an α- or β-decay (daughter nucleus in an excited state)
- $T_{½}$ is generally small ($< 10^{-9}$ s) but sometimes can be $\gg$ (isomeric or metastable states) → no clear distinction between states which are isomeric or not → 10^{-6} s is isomeric and 10^{-12} s is not → in between: fuzzy
- Competing process → internal conversion: the energy is transferred to an atomic electron (no *Z* and *N* changes for the nucleus but the atom becomes ionized)

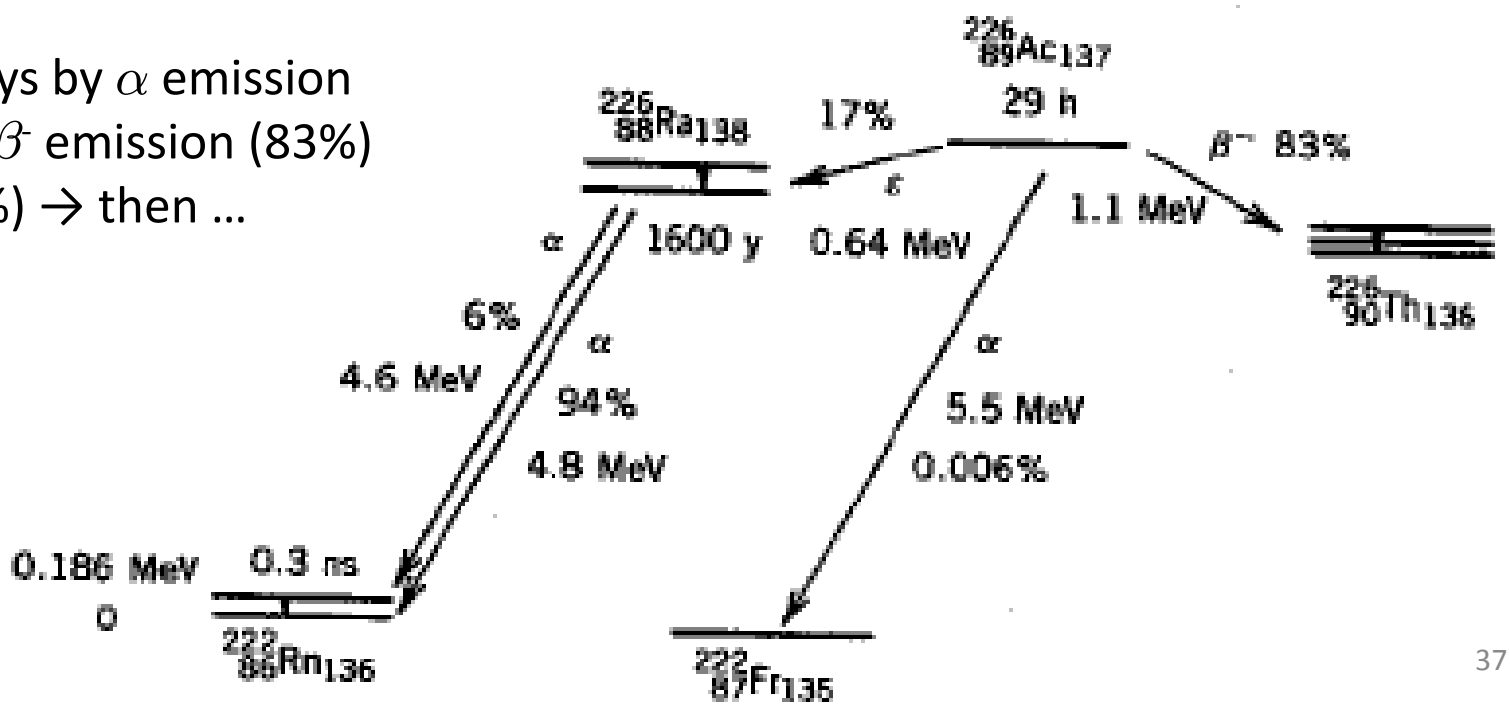
Other processes

- **Spontaneous fission**: some nuclei spontaneously fission → a heavy nucleus with an excess of neutrons splits roughly in half into two lighter nuclei → the final nuclei are not rigidly determined but are statistically distributed over the entire range of medium-weight nuclei + neutrons + γ-rays +…(examples: ^{256}Fm with $T_{½}$ = 2.6 h or ^{254}Cf with $T_{½}$ = 60.5 days)
- **Nucleon emission**: As we move further and further from the stability valley → the energy differences between neighboring isobars ↗ → can be larger than the nucleon binding energy (≈ 8 MeV) →radioactive decay by nucleon emission → occurs most frequently in fission products having a large neutron excess → delayed neutrons → very important in the control of nuclear power plants (ex: $^{138}\text{Xe} \rightarrow {}^{137}\text{Xe} + \text{n}$ or $^{73}\text{Br} \rightarrow {}^{72}\text{Se} + \text{p}$)

Branching ratios (1)

- Often it exists several possible decay processes in competing mode → the decay schemes may be very complicated
- We specify the relative intensities of the competing modes by their branching ratios → example:

^{226}Ac decays by α emission (0.006%), β^- emission (83%) and ϵ (17%) → then …



Branching ratios (2)

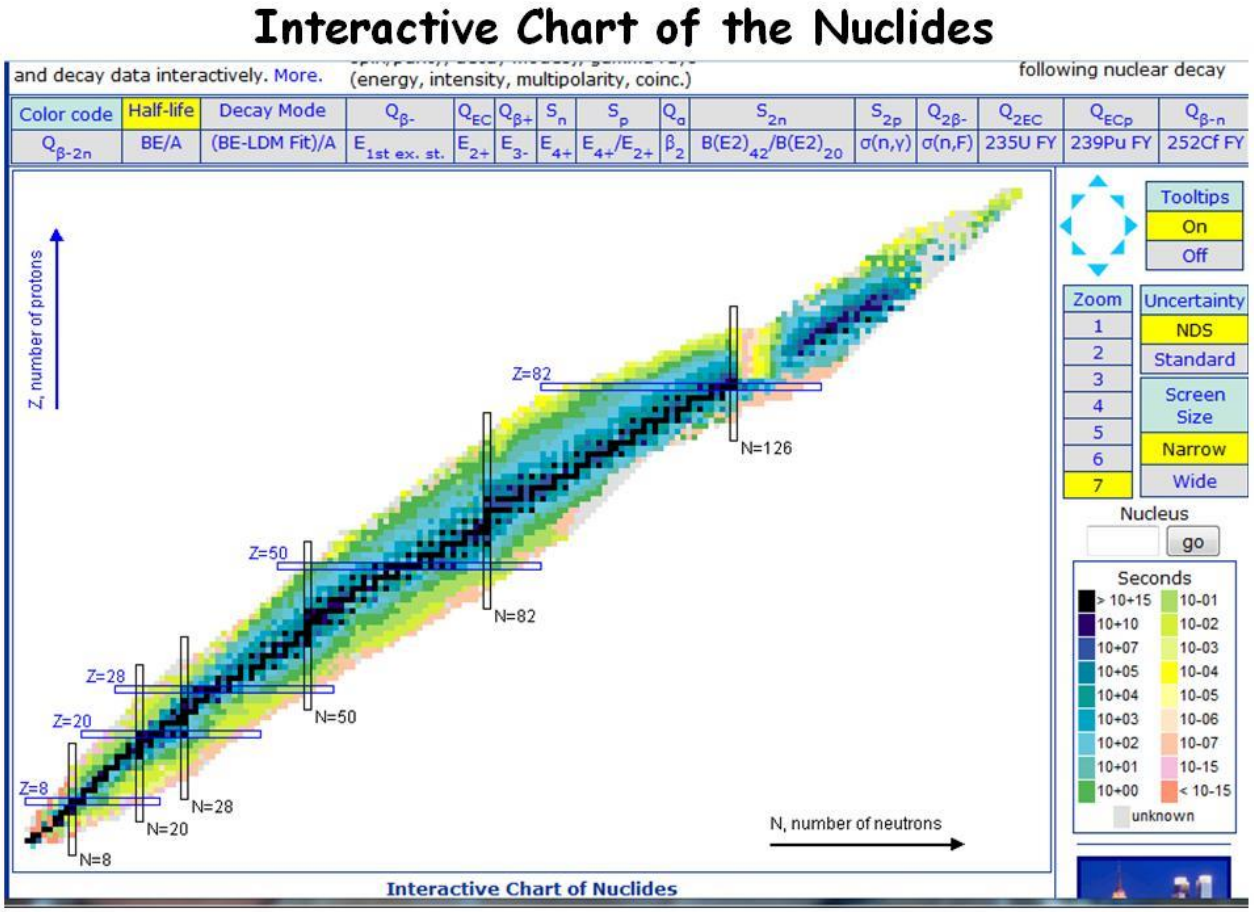
- Frequently → branching ratio is specified by giving the partial decay constant or partial half- life
- For ^{226}Ac →

$$\begin{aligned}
\lambda_t &= \frac{0.693}{29h} = 0.024h^{-1} = 6.6 \times 10^{-6}s^{-1} \\
\lambda_\beta &= 0.83\lambda_t = 5.5 \times 10^{-6}s^{-1} \\
\lambda_\epsilon &= 0.17\lambda_t = 1.1 \times 10^{-6}s^{-1} \\
\lambda_\alpha &= 6 \times 10^{-5}\lambda_t = 4 \times 10^{-10}s^{-1}
\end{aligned}$$

- Partial half-life ($T_{½,I}$ = 0.693/λ_i) are convenient to represent branching ratio → **but only total half-life has a sense**

interactive chart of nuclides

- All information → interactive chart of nuclides: **http://www.nndc.bnl.gov/chart**



Summary of various decay processes